CHANGE CYCLICITY OF VOLUMETRIC ACTIVITY OF RADIONUCLIDE ^{137}Cs IN COASTAL WATERS OF THE BALTIC SEA AND ITS POSSIBLE REASONS

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Abstract. Monitoring of the volumetric activity (VA) of a radionuclide ^{137}Cs in the coastal waters of the Baltic Sea was carried out near the settlement of Juodkrantė in the summer – autumn of 2004. Three measurement stages of the VA of this radionuclide were carried out and hydrometeorological situations were considered simultaneously. ^{137}Cs VA change cyclicity was found out near the seacoast. Extreme data for the whole period of observation were 48 and 94 Bq/m^3. Maximal values exceeded minimal data approximately two times, which practically coincided with the radioactive "background". The method of mathematical modeling was used to find out possible reasons of ^{137}Cs VA change near the seacoast. Because of lack of information about the location and power of a possible "source" of radioactive pollution, repeated calculations were carried out, assuming its various places of formation and various power, i e search of optimum coincidence of calculation and measurement data was selected. Optimum coincidence of theoretic and measurement results was found at the location of a radioactive "source" in the southern part of the Lithuanian sea area at a distance of 25–35 km from Juodkrantė. The VA of the radioactive "source" was 5000 Bq/m^3 in the summertime and 15000 Bq/m^3 in October of 2004. Agreement of measurement and calculation results was satisfactory for all the observation stages.

Keywords: the Baltic Sea, radionuclide ^{137}Cs, comparison, mathematical modeling, radioactive "source".

1. Introduction

It is known that artificial radionuclides have penetrated into the Baltic Sea from the global fallout and radioactive waste discharge [1]. In 1986 large amounts of artificial radionuclides penetrated after the Chernobyl Power Plant (ChPP) accident too [1].

The fallout of radionuclides on the surface of the Baltic Sea was heterogeneous, therefore, the leveling process of their volumetric activity (VA), including ^{137}Cs, occured during a long period of time. Up to now the waters of the Baltic Sea are contaminated by this radionuclide. However, high values of ^{137}Cs VA in the surface waters were marked from 1988 to 1995 [2]. Possible reason of such a situation was an influence of the waters of the Gulf of Bothnia where a higher VA of ^{137}Cs was observed [3]. Later the VA of a radionuclide ^{137}Cs considerably decreased, however, during the self-purification process there were some exceptions as, for example, in 1999 and 2002, when the VA values increased [4].

Comparison of average annual values of the measurement and calculation data of the radionuclide ^{137}Cs VA were not in good agreement, and calculated values appeared below measurement results.

It is necessary to note that at rather a small number of measurements and their results averaging the course of ^{137}Cs VA in the southeastern part of the Baltic Sea and its coastal waters near Juodkrantė in 1996–2001 appeared approximately the same [3].

However, the results of accidental measurements in various parts of the Baltic Sea can be different. On the one hand it can be connected with a change of hydrometeorological situations, on the other hand – with an additional radioactive waste discharge [5]. Random measurement results of radionuclide ^{137}Cs VA have an accidental character; for example, it took place in 1998, 1999, 2002 [6]. To define the regularity of the course of ^{137}Cs VA in coastal waters of the Baltic Sea long-term measurements are necessary. To realize this monitoring in practice the observation station of Juodkrantė was chosen. Three stages of measurement were carried out in the summer – autumn period of 2004. This monitoring contributed to a detailed analysis of the VA course of ^{137}Cs near the seashore. On the basis of mathematical modeling possible cause of the obtained regularity was set up. The above stated aspects define the aim of the present work.

2. Measurement technique

The present study used the procedure of ferrocyanide-carbonate precipitation. A radionuclide ^{137}Cs was precipitated from seawater samples of 40–50 l in volume [5–7]. Water sampling was carried out at a depth of 0,5 m in coastal waters.

The yield of Cs was determined gravimetrically in the form of $Cs_3Sb_2I_9$. The yield value varied within 60–80 %. The activity of ^{137}Cs samples was registered by a gamma spectrometer with a semiconductor detector. The determination error for ^{137}Cs VA amounted to 10 %.

3. Results and analysis

It is known that the hydrosphere has been contaminated with artificial radionuclides from the global fallout since 1954, when for the first time a radionuclide ^{90}Sr was found in the Atlantic Ocean. Increase of the global fallout proceeded up to 1963, when a treaty concerning nuclear and thermonuclear weapon test stop in the atmosphere, hydrosphere and cosmos was signed. This moment was the beginning of decrease in radioactive contamination of the environment. This process was observed up to 1986, i e up to the ChPP accident.

After this accident the radioactivity of the environment considerably increased, including a radionuclide ^{137}Cs in the surface waters of the Baltic Sea. The real self-purification process of these waters began in 1989, when an approximately identical VA of this radionuclide was observed in the whole area of the Baltic Sea [8].

According to the data obtained by a theoretical model, an average value of the VA of a radionuclide ^{137}Cs has to be 40–50 Bq/m^3 at the present time [9]. This value is in good agreement with the obtained results in the southeastern part of the Baltic Sea and lower than those in its coastal waters [6].

However, the results of accidental measurements in some cases differ considerably from average values (Table 1).

Table 1. Values of volumetric activity (VA) of radionuclide ^{137}Cs and some hydrometeorological parameters in coastal waters of the Baltic Sea near Juodkrantė settlement in 2004

No	Date	Temperature, °C	Wind direction	Average wind velocity, m/s	^{137}Cs VA, Bq/m^3
1	30 Jun	16	West	3	66 ± 7
2	1 Jul	16	West	4	60 ± 6
3	2 Jul	15	West	4	53 ± 5
4	3 Jul	16	West	3	54 ± 5
5	4 Jul	17	South	3	59 ± 6
6	5 Jul	17	West	5	58 ± 6
7	6 Jul	16,5	West	6	57 ± 6
8	7 Jul	17	West	5	55 ± 6
9	8 Jul	17,5	South	2	61 ± 6
10	9 Jul	17,5	East	6	48 ± 5
11	10 Jul	17	West	5	50 ± 5
12	12 Jul	16	South	7	57 ± 6
13	13 Jul	16,5	West	5	80 ± 8
14	14 Jul	16	West	5	84 ± 8
15	15 Jul	16	West	3	74 ± 7
16	17 Jul	16	West	3	94 ± 9
17	18 Jul	17	East	2	77 ± 8
18	24 Aug	17	East	3	64 ± 6
19	26 Aug	17,5	South	2	76 ± 8
20	28 Aug	17	South	5	91 ± 9
21	29 Aug	17,5	South	3	76 ± 8
22	30 Aug	18	South	3	57 ± 6
23	1 Sept	17,5	South	5	54 ± 5
24	2 Sept	17,5	West	5	50 ± 5
25	3 Sept	17	Northwest	6	49 ± 5
26	4 Sept	18	North	3	52 ± 5
27	5 Sept	18	West	1	55 ± 6
28	16 Oct	9,5	East	5	60 ± 6
29	18 Oct	10	Southeast	5	70 ± 7
30	20 Oct	10,5	North	1	65 ± 6
31	22 Oct	10	Southwest	6	72 ± 7
32	24 Oct	11,5	West	5	71 ± 7
33	26 Oct	11	West	5	70 ± 7
34	28 Oct	10	East	3	52 ± 5
35	30 Oct	10	East	4	55 ± 6
36	16 Jul (Nida)	16	West	3	72 ± 7
37	16 Jul (Pervalka)	16	West	3	71 ± 7
38	4 Sept (Nida)	17	North	3	69 ± 7
39	4 Sept (Preila)	18	North	3	70 ± 7
40	4 Sept (Pervalka)	18	North	3	55 ± 6

According to the data of Table 1, the values of ^{137}Cs VA did not fall outside the limits of experimental errors from 30 Jun to 12 Jul. However, since 13 Jul an increase of ^{137}Cs VA with the greatest value on 17 Jul (94 Bq/m^3) was observed. An increase of ^{137}Cs VA at the other stations of the Lithuanian seacoast – Pervalka and Nida – was also registered.

The next leap of ^{137}Cs VA (91 Bq/m^3) was observed on 28 Aug, and since 30 Aug the values of VA were close to the radioactive "background" (50–60 Bq/m^3). An increase of ^{137}Cs VA was also observed on 22–26 Oct. Raised values, as a rule, were registered during the wind direction change. In particular, in the first case (12–13 Jul) the southern wind was replaced by the western wind on 24–28 Aug, the eastern wind was replaced by the southern wind, and on 20–24 Oct the northern wind was replaced by the western wind (Table 1).

Within July – September an identical temperature at the seashore and rather a small velocity of the wind was observed. However, the extreme values of ^{137}Cs VA were 48 and 94 Bq/m^3 for this period, i e their ratio was equal to 1:2. Such leaps of ^{137}Cs VA cannot arise only owing to hydrometeorological situations [5]. The most probable cause of the above mentioned leaps is an additional radionuclide ^{137}Cs discharge to the Baltic Sea. As these leaps are a frequent phenomenon in the coastal zone [5], a theoretical investigation is necessary.

Only a great number of measurement results can find out the regularity of leap formation cyclicity of ^{137}Cs VA in the coastal waters of the Baltic Sea. Unfortunately, parallel observations in the open part of the sea were impossible during summer – autumn monitoring because of lack of technical means.

4. Theoretic model of passive admixture transfer in the Baltic Sea

For a theoretic substantiation of leap formation of ^{137}Cs VA in the coastal waters of the Baltic Sea, a mathematical model was tested and proposed. The basic aspects of the model are presented below.

Distribution of the concentration of a passive admixture in the surface waters of the sea area at the presence of a dot source, that is located at a certain point of this area with co-ordinates (x_s, y_s), is described by the equation of diffusion:

$$\frac{\partial C}{\partial t} = \operatorname{div}\left(K \operatorname{grad} C\right) - \vec{v} \operatorname{grad} C, \qquad (1)$$

where $C = C(x, y, t)$ – concentration of an admixture, $\vec{v} = v(x, y)$ – vector of velocity of water mass motion, K – coefficient of turbulent diffusion.

To solve equation (1) the following boundary and initial conditions are accepted. On coastal line Γ_C of area – a condition of nonflowing of admixture is used:

$$\left.\frac{\partial C}{\partial n}\right|_{\Gamma_c} = 0,$$

where n – direction of a normal to coast. On the remaining part of the contour of water area Γ_w, because of its remoteness from a source and a short term of admixture distribution, the condition of a constant concentration is accepted:

$$C|_{\Gamma_w} = C_f,$$

where C_f – "background" concentration. It is also supposed that initial distribution of the concentration of a passive admixture and the background concentration are the same:

$$C(x, y, 0) = C_f.$$

At all the points of the area except the spot of the source location

$$C(x_s, y_s, 0) = C_S,$$

where C_S – concentration of a passive admixture of a "source".

For a numerical solution of the problem, various schemes of solution of two-dimensional diffusion were checked. However, because of rather a slowed down levelling process of the concentration of an admixture in the seawater, an obvious scheme with a unilateral approximation [10] was chosen:

$$C_{i,j}^{n+1} = C_{i,j}^n +$$

$$+ \frac{K\Delta t}{h^2}\left(C_{i-1,j}^n + C_{i+1,j}^n + C_{i,j-1}^n + C_{i,j+1}^n - 4C_{i,j}^n\right) -$$

$$- \frac{\Delta t}{h}\lambda u_{i,j}\left(C_{i,j}^n - C_{i-\lambda,j}^n\right) + \mu v_{i,j}\left(C_{i,j}^n - C_{i,j-\mu}^n\right),$$

where

$$\lambda = \begin{cases} +1, u > 0 \\ -1, u < 0 \end{cases} \qquad \mu = \begin{cases} +1, v > 0 \\ -1, v < 0 \end{cases}.$$

Such a scheme has a high stability, its realization is simply programmed and also proceeds with a high velocity. For calculating the field of velocity $v = v(x, y)$, a stationary model [11] was used:

$$\operatorname{div}\left(\frac{1}{H^3}\operatorname{grad}\psi\right) = \frac{1}{2a}\operatorname{rot}_z\left(\frac{T}{H}\right),$$

$$\psi|_{\Gamma_w} = 0, \qquad (2)$$

$$\left.\frac{\partial \psi}{\partial n}\right|_{\Gamma_w} = 0,$$

where $\psi = \psi(x, y)$ – function of full streams, α – coefficient of vertical exchange, $T = \{T_x, T_y\}$ – vector of tangential wind tensity, H – depth of the sea. The velocity of surface water motion is calculated by the formulas:

$$u = H\frac{T_x}{4a} - \frac{3}{2H}\frac{\partial \psi}{\partial y},$$

$$v = H\frac{T_y}{4a} + \frac{3}{2H}\frac{\partial \psi}{\partial x}.$$

Numerical realization of diffusion (1) and advection (2) problems was carried out on the same net area (x_i, y_j) with a step of $h = 4$ km. Therefore, after calculating the field of velocities, the problem of admixture spreading with a conclusion to the screen of isolines of the concentration of a passive admixture was solved.

A theoretical model was applied for calculating the structure of the VA field of a radionuclide ^{137}Cs in a southeastern and coastal area of the Baltic Sea. As the authors knew only hydrometeorological situations, and

there was no information on the location and power of an imaginary radioactive source, selection of the latter characteristics was necessary. These characteristics were chosen in such a way that the calculation results would coincide with the measurement data in the range of an experimental error. It is natural that an unequivocal solution of such a problem is practically impossible because of a large number of initial and boundary conditions. Hence, the location and power of a "source" were set until the coincidence of calculation and measurement data was optimum.

The background of ^{137}Cs VA value reached 50 Bq/m^3 [9]. The calculation results were compared with the measurement data at the coast of the Baltic Sea according to Table 1. The main attention was paid to that situation only when the measurement results of ^{137}Cs VA had the greatest values.

It is found out [12] that a moving "spot" of a passive admixture had a maximum velocity towards the shore in the western and southwestern directions of the wind, when an increase of ^{137}Cs VA was observed. During the measurements of ^{137}Cs VA in 2004 the above stated situations were observed three times, however, the wind velocities were lower than in paper [12].

The calculation results of a real ^{137}Cs VA in the coastal waters of the Baltic Sea were possible, when the initial location of a conditional radioactive "source" had the co-ordinates x = 51, y = 71 (Fig 1) at a distance of about 33 km from the settlement of Juodkrantė with ^{137}Cs VA of 5000 Bq/m^3. This position extended to a real situation which occurred on 10–17 Jul (Fig 2, 3). Here the calculation and measurement results were in satisfactory agreement in the western and southwestern directions of the wind with an average velocity of 5 m/s. However, this agreement appeared to be unsatisfactory on 18 Jul because of a change of the wind direction (Table 1). This fact was not considered with a theoretical model in this case.
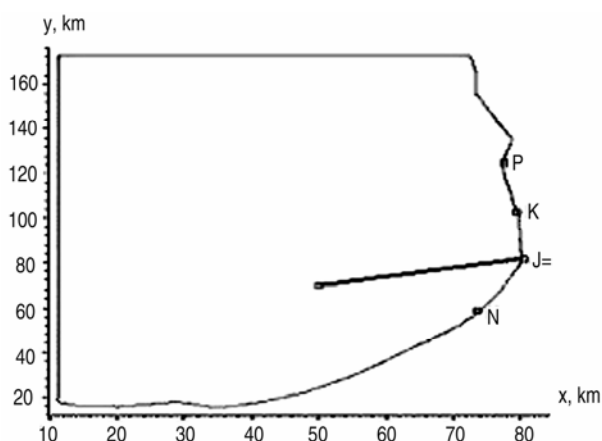

Fig 1. Location of imaginary radioactive (radionuclide ^{137}Cs) "source" in surface waters of Lithuanian marine area with power of 5000 Bq/m^3 on 10 Jul 2004. P – Palanga; K – Klaipėda; J – Juodkrantė; N – Nida

It is necessary to point out that Figs 2 and 3 (a–h) also illustrate dispersion of a conditional radioactive "spot". It appeared that in an initial place of formation of this "spot" of ^{137}Cs VA after 8 days it decreased from 5000 to 60 Bq/m^3. Comparison of the measurement and calculation results of ^{137}Cs VA are presented in Fig 4.

Here the majority of compared results does not fall outside the limits of experimental errors. Measurement (Table 1) and calculation (Fig 5) results illustrate an increase of ^{137}Cs VA in other stations of the Lithuanian seashore. Here the least values fall to the city of Palanga (Fig 5a), as this point is at the largest distance from a "source" of pollution.
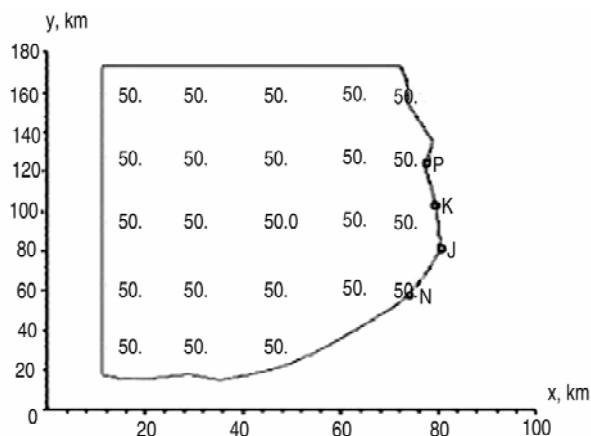

Fig 2. Initial structure of ^{137}Cs VA field (Bq/m^3) in Lithuanian marine area on 10 Jul 2004

Taking into consideration similar hydrometeorological conditions, the location of a radioactive "source" and the initial VA of 10000 Bq/m^3, the calculation results are in bad agreement with the measurement data near the coast of Juodkrantė. In fact the measurement value was 94 Bq/m^3 and the calculation result was 136 Bq/m^3 on 17 Jul. It means that the present analysis gives possibility to assess the VA of an accidental "source".

Satisfactory agreement of the theoretical and experimental data occured from 24 Aug to 5 Sept, when the extreme results were 49 and 91 Bq/m^3, i e they were almost the same as on 10–18 Jul (Table 1), and had the same VA of a "source" (5000 Bq/m^3).

Leap of ^{137}Cs VA was lower in Oct than in the summertime (Table 1). During the measurements on 16–30 Oct change of the wind direction was observed twice that led to change of ^{137}Cs VA near the seacoast. In these cases the mathematical model considered changes of hydrometeorological situations. Change of the wind direction was observed on 19–20 Oct and on 28 Oct.

The location of a "source" was set at a point of the sea with co-ordinates x = 59 and y = 71 for the initial VA of 15000 Bq/m^3. Comparison of measurement and calculation data is presented in Table 2.

Here change in the direction and velocity of the wind (Table 1) influenced the value of ^{137}Cs VA and reduced it up to the "background" value on 28 Oct.
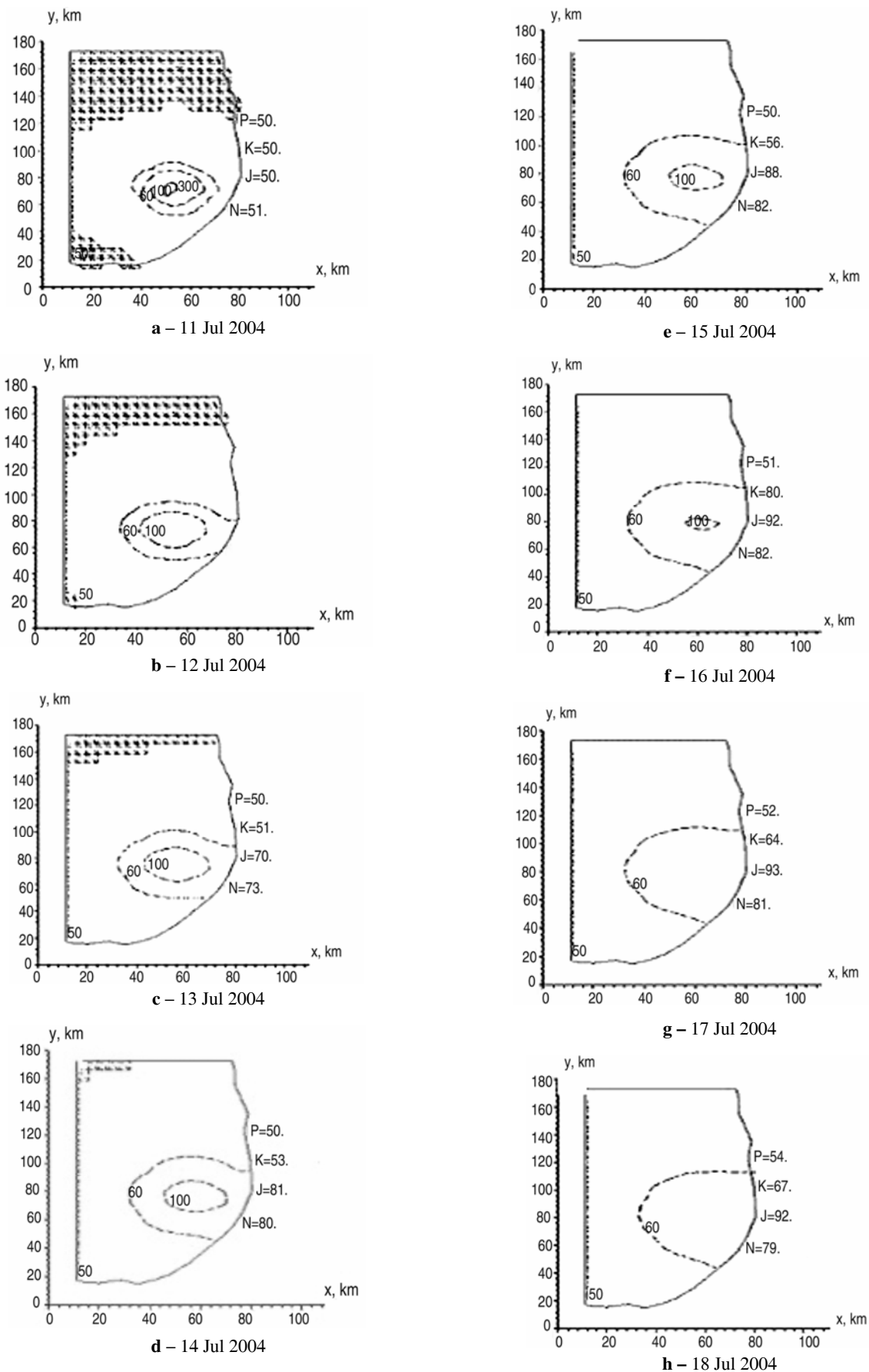
Fig 3 (a, b, c, d, e, f, g, h). Structure of ^{137}Cs VA (Bq/m^3) field in Lithuanian marine area. Numbers next to letters P, K, J, N mean ^{137}Cs VA (Bq/m^3) at mentioned points (Fig 1)
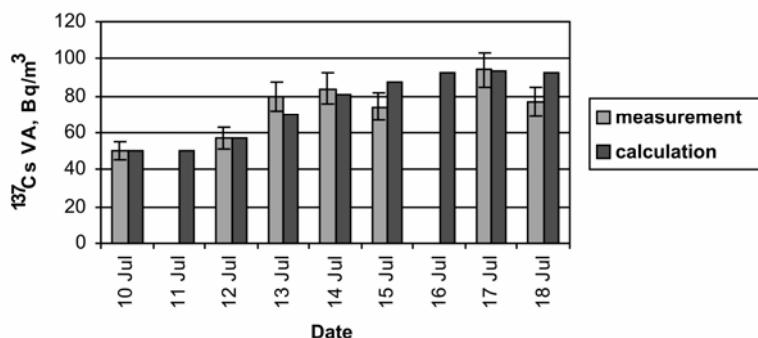
Fig 4. Comparison of measurement and calculation data of ^{137}Cs VA (Bq/m^3) in coastal waters of the Baltic Sea near Juodktantė settlement on 10–18 Jul 2004

Table 2. Measurement and calculation values of ^{137}Cs VA (Bq/m^3) in coastal waters of the Baltic Sea near Juodkrantė settlement in Oct 2004

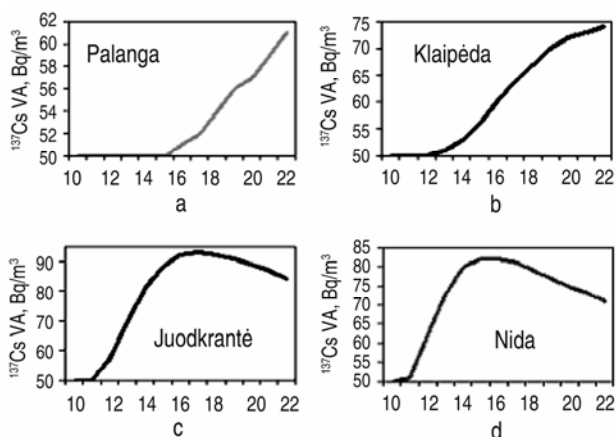| 1	Date	14 Oct	16 Oct	18 Oct	20 Oct	22 Oct	24 Oct	26 Oct	28 Oct	30 Oct
2	Experiment	–	60	70	65	72	71	70	52	55
3	Theory	50	60	67	62	68	69	60	51	50

Fig 5. Calculation results of 137Cs VA (Bq/m3) change at points of Lithuanian seashore (Fig 1) from 10 Jul 2004 to 22 Oct 2004

5. Discussion

The measurements of ^{137}Cs VA in the coastal waters of the Baltic Sea near the settlement of Juodkrantė often showed regularity of the course of its values in 2004. Regular leaps of ^{137}Cs VA and its decrease up to the background values were found out.

Unfortunately, the calculation model cannot consider in detail the changes of hydrometeorological parameters and resulting change of water mass transfer. It is obvious that if information of a "source" location and its power are unknown, theoretical results are considered to be only approximate.

However, a theoretical model confirms probability of the existence of such a "source". It is obvious that a high ^{137}Cs VA (91–94 Bq/m^3) near the seashore would be impossible without this "source".

Unfortunately, continuous control over the VA of ^{137}Cs in the whole Lithuanian sea area for a long time is impossible because of lack of technical means. Therefore, the situation can be controlled theoretically, confirming the results of calculations only by regular experimental data in the coastal zone.

It has appeared that the most probable place of formation of a radioactive "source" with an increased value of ^{137}Cs VA is in the southern part of the Lithuanian marine area, and the imaginary "source" is located at a distance of 25–35 km from an observation station at the settlement of Juodkrantė.

It is fixed that a rapid change of the VA of a radionuclide ^{137}Cs in surface waters is influenced by change of the wind direction as well as the direction of the sea current. It means that the radionuclide VA is heterogeneous in surface waters, and a change of the sea current direction leads to radionuclide VA change.

The slowing down process of the self-purification of the Baltic Sea waters is probably connected with formation of local radioactive "sources". Their origin can depend on different reasons analysis of which is presented in this work.

It is necessary to note that in the above schemes of the southeastern part of the Baltic Sea real geographical co-ordinates are not presented, and the location of an imaginary "source" is defined by conditional co-ordinates because of a low accuracy of its detection. It is obvious that increase in the number of observations at several points of the coastal zone would allow to find the location of a "source" with a greater accuracy and calculate its power.

6. Conclusions

1. Cyclic regularity of formation of increased values of ^{137}Cs VA in the coastal waters of the Baltic Sea near the settlement of Juodkrantė is found out.

2. Extreme experimental values of a radionuclide ^{137}Cs were 48 and 94 Bq/m^3 in the summer – autumn of 2004. Such a difference of the obtained results could not be caused only by change of natural hydrometeorological situations.

3. The presented theoretical model defines distribution of a passive admixture in coastal waters of the Baltic Sea under any hydrometeorological situations.

4. Adjustment of theoretical results according to experimental data using statistical material proves the existence and approximate location of a possible radioactive "source" in the Lithuanian marine area.

The mathematical model turned out to be the most efficient means of obtaining reliable information on ^{137}Cs VA at any point of the marine area under stationary hydrometeorological situations.

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RADIONUKLIDO ^{137}Cs TŪRINIO AKTYVUMO KITIMO CIKLIŠKUMAS BALTIJOS PAKRANTĖS VANDENYSE IR JO GALIMOS PRIEŽASTYS

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S a n t r a u k a

2004 m. vasarą – rudenį atliktas radionuklido ^{137}Cs tūrinio aktyvumo (TA) stebėsena Baltijos jūros pakrantės vandenyse prie Juodkrantės. Radionuklido TA matavimai vykdyti trimis etapais. Tuo pačiu metu buvo įvertintos hidrometeorologinės situacijos.

Nustatytas ^{137}Cs TA kitimo pakrantės vandenyse cikliškumas. Minimali ir maksimali TA reikšmės per visą matavimo periodą buvo atitinkamai lygios 48 ir 94 Bq/m^3. Mažiausiosios reikšmės apytikriai 1,5–2,0 kartų viršijo didžiausiąsias, kurios iš esmės sutapo su radioaktyviuoju „fonu".

Galimoms ^{137}Cs TA kitimo pakrantės vandenyse priežastims nustatyti taikytas matematinio modeliavimo metodas. Nesant informacijos apie galimo šaltinio vietą ir jo galingumą, atlikti daugkartiniai skaičiavimai. Skaičiuojant buvo keičiama šaltinio vieta ir galingumas, t. y. siekta nustatyti matavimo prie Juodkrantės ir skaičiavimo rezultatų optimalų sutapimą. Geriausias teorinių ir eksperimentinių duomenų sutapimas gautas, esant radioaktyviajam „šaltiniui" Lietuvos jūrinės zonos pietinėje dalyje 25–35 km atstumu nuo Juodkrantės, kai vasarą jo TA lygus 5 000 Bq/m^3, bei 2004 m. spalio mėnesį – 15 000 Bq/m^3. Apskaičiuotieji duomenys su gautaisiais visų stebėjimo serijų metu sutampa patenkinamai.

Prasminiai žodžiai: Baltijos jūros pakrantės vandenys, radionuklidas ^{137}Cs, palyginimas, matematinis modeliavimas, radioaktyvusis „šaltinis".

ЦИКЛИЧНОСТЬ ХОДА ОБЪЕМНОЙ АКТИВНОСТИ РАДИОНУКЛИДА ^{137}Cs В ПРИБРЕЖНЫХ ВОДАХ БАЛТИЙСКОГО МОРЯ И ЕГО ВОЗМОЖНЫЕ ПРИЧИНЫ

Д. Стыро, Й. Клейза, Р. Моркунене, А. Даунаравичене

Р е з ю м е

Осуществлен мониторинг объемной активности (ОА) радионуклида ^{137}Cs в прибрежных водах Балтийского моря у пункта наблюдений – поселка Юодкранте летом–осенью 2004 г. Выполнены три серии измерений этого радионуклида. Одновременно учитывались гидрометеорологические ситуации.

Обнаружена цикличность хода ОА ^{137}Cs у берега моря. Экстремальными данными за весь период наблюдений оказались 48 и 94 Бк/м^3. Максимальные значения примерно в 1,5–2,0 раза превосходили минимальные, которые практически совпадали с радиоактивным „фоном".

Для выяснения возможных причин хода ОА ^{137}Cs у берега моря применялся метод математического моделирования. Из-за отсутствия информации о местонахождении и мощности возможного источника радиоактивного загрязнения проводились многократные расчеты. При этом предполагались разные места его образования и разная мощность, т. е. проводился поиск оптимального совпадения расчетных данных и данных измерений.

Теоретические и экспериментальные результаты, когда ОА в летнее время было 5000 Бк/м^3 и в октябре 2004 г. – 15000 Бк/м^3, лучше всего совпадали, когда радиоактивный источник находился в южной части литовской морской зоны на расстоянии 25–35 км от Юодкранте. Совпадение сопоставляемых величин оказалось удовлетворительным для всех серий наблюдений.

Ключевые слова: Балтийское море, радионуклид ^{137}Cs, сравнение, математическое моделирование, радиоактивный источник.

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